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I. A. Pawelczak, A. M. Glenn, H. P. Martinez, M.
L. Carman, N. P. Zaitseva, S. A. Payne

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Boron-loaded Plastic Scintillator with Neutron- γ Pulse Shape Discrimination Capability

I.A. Pawełczak, A.M. Glenn, H.P. Martinez, M.L. Carman, N.P. Zaitseva,
S.A. Payne

Lawrence Livermore National Laboratory, Livermore, CA 94550

Abstract

Development of plastic scintillator with neutron sensitivity from thermal to multi-MeV and pulse shape discrimination (PSD) has been demonstrated. Incorporation of ^{10}B -containing compounds into plastic scintillator with PSD capability leads to detector improvement in regards to neutron detection efficiency while preserving the discrimination between neutrons and γ -rays. Effects of boron loading on scintillation and pulse shape discrimination properties are discussed. PSD figure-of-merit value of 1.4 ± 0.03 has been achieved for events in thermal neutron energy domain, 50-100keV_{ee}, for PSD plastic loaded with 5wt.% of m-carborane.

Keywords: ^{10}B -loaded PSD plastic scintillator, Pulse shape discrimination, Thermal-neutron detection, Fast-neutron detection

1. Introduction

Neutron count rates, multiplicity and energy distributions serve as an indicator for the presence of Special Nuclear Material (SNM). Therefore, efficient solid-state neutron detectors with fast response and large active volume are in high demand for neutron detection in nuclear security and safeguards applications. Conventional technologies for neutron detection are based on ^3He for thermal-neutron detection, and liquid scintillators for fast-neutron detection. However, due to the shortage of ^3He supplies and the difficulty of handling and safety issues associated with liquid scintillators, new alternatives are under consideration. In addition, field deployment imposes a

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stringent requirement on detector operation in a self-triggering mode, and therefore detector internal ability to identify and distinguish neutrons from a γ -rays.

Studies of pulse shape discrimination phenomena in organic crystals and liquids led to the development of plastic scintillators with pulse shape discrimination (PSD) [1]. Incorporation of efficient thermal neutron converters such as ^{10}B or ^6Li into plastic scintillators with PSD capability increases neutron detection efficiency while discriminating neutrons from γ -rays on an event-by-event basis. Recent research has shown that ^6Li -loaded PSD plastics provide discrimination among thermal neutrons, fast neutrons and γ -rays [2], however the larger ^{10}B thermal neutron cross-section and ease of loading allow for significantly higher thermal neutron capture efficiency than for ^6Li -loaded plastic of the same size. Nonetheless, boron loaded PSD plastics are best suited to smaller size detectors due to the lower reaction Q-value, so ^6Li and ^{10}B loaded PSD plastics represent complementary technologies. In this manuscript, the sensitivity of PSD plastic scintillator to thermal neutrons is achieved by using the capture reaction on a nucleus of ^{10}B , according to [3]:



^{10}B loading was selected due to the high cross-sections for thermal neutron capture (72% of ^3He), high natural abundance (19.8%), emission of light charged particles in thermal neutron capture reaction, and chemical compatibility with plastic formulation. It is known that charged reaction products, (^7Li and ^4He), require only several tens of μm to be stopped in the scintillator. Their short range is desired for achieving good efficiency in a reasonable size detector. The ^7Li and ^4He products also have significantly different stopping power, dE/dx , from minimum ionizing particles such as recoiled electrons (γ -rays) which enhances PSD.

A boron-containing PSD plastic detects fast neutrons by recoiled protons and thermal neutrons by capture reaction products while discriminating against background γ -rays. The scintillator serves dual purposes as a detector and neutron moderator due to its high hydrogen content. This allows for accounting of neutrons which are below the detection threshold by recoiled protons and above the effective capture by thermal neutron converters. Neutrons thermalized within the detector volume diffuse until captured by ^{10}B or escape from the detector. This approach may allow for efficient detection

of fission neutrons with a compact detector design where external moderator is not required.

Moreover, detection of a multi-MeV neutron by a recoiled proton followed by detection of the capture reaction products within the scintillator provides additional information. Time correlated signals, fast generated due to recoiled proton, and diffusion delayed due to neutron capture, ensure neutron full energy deposition to the scintillator and provide improved neutron spectroscopy [4, 5, 6, 7]. Such a detector may be also considered to provide multiplicity information for neutron bursts [8] in a cost-effective way.

Organic liquid scintillators containing ^{10}B compounds have been used in the past for thermal neutron detection [9, 10]. High flash point liquid scintillators with PSD [11, 12, 13, 14, 15] were investigated for border monitoring applications and reported efficiency comparable to ^3He counters, although with significantly lower γ -ray rejection levels. On the other hand, solid state scintillators containing natural boron (glass, plastic and silicon rubber based) have been considered mainly as materials without PSD capability [16, 17, 18]. Existing boron-containing plastic scintillators without PSD use energy deposition, pulse height (PH), specific to charged reaction products, to identify neutrons. Such scintillators would benefit from the addition of PSD to provide additional rejection of omnipresent γ -rays events underneath the thermal neutron PH distribution.

The work presented in this paper is focused on the development of PSD plastic scintillator loaded with ^{10}B . Such a detector provides sensitivity to neutrons in a wide dynamic range and pulse shape discrimination capability. Furthermore, influences of m-carborane concentration, a compound used to introduce boron into plastic, on scintillator light emission properties and pulse shape discrimination are reported.

2. Experimental Methods and Simulation Calculations

Samples of PSD plastic containing varied concentrations of boron were made as colorless, optically transparent scintillators by dissolution of m-carborane, 28% of PPO (2,5-diphenyloxazole) and 0.2% of DPA (9,10-diphenylanthracene) into vinyltoluene or styrene. Carborane was used as a boron-containing compound because of its highest concentration of boron atoms per molecule among all known compounds compatible with organic scintillators. Natural abundance of boron in m-carborane was used due to significantly lower cost than

its enriched form. Performance tests of synthesized plastic scintillator samples with respect to neutrons were carried out in the presence of γ -rays using a ^{252}Cf source. In order to attenuate the flux of γ -rays and moderate neutrons, 5-cm of Pb and 7.5-cm high density polyethylene, HDPE, were placed between the scintillator and the source. Charge integration was used as a discrimination method where the difference in the amount of delayed light generated by proton and electron recoil is used to identify neutron and γ -ray events.

Experiments were conducted with scintillator samples of 2.5-cm diameter and 1-cm thickness. Scintillators were coupled to a Hamamatsu 2-inch R6231-100 photomultiplier with photocathode maximum sensitivity at 420nm. This PMT is well matched to the wavelength of maximum light emission for the developed scintillators. Signals from the E1198-27 photomultiplier base were transmitted to a 14-bit, 200 MHz 14200Gage Compuscope digitizer. Full waveforms were collected and analyzed offline. Energy calibration was performed using 662keV γ -rays from ^{137}Cs source assuming Compton edge energy of 478keV_{ee} at 90% height of Compton edge distribution. PSD optimization was performed for events in the 50-100keV_{ee}, the energy range in which thermal neutron signature is presented with the highest intensity. The equation for the PSD is given by:

$$PSD = \frac{\sum_{del_{start}}^{del_{end}} Qdt}{\sum_{tot_{start}}^{del_{end}} Qdt}, \quad (2)$$

where del_{start} and del_{end} are respectively the beginning and end of the gate used for integration of charge generated due to delayed light, while tot_{start} corresponds to the beginning of the gate used for integration of total (prompt and delayed) charge. The beginning of the gate used for total charge integration was fixed at fourteen samples (70 ns) before the maximum peak amplitude. Integration of the delayed component was started 9 samples (45 ns) from pulse maximum and proceeded for 250 samples (1250 ns) to achieve the highest separation of neutron and γ -ray events in the energy range characteristic to thermal neutron capture. The integrated charge was corrected for the baseline, averaged over eighty pre-trigger samples. To quantify PSD, 1-D PSD distributions in energy bin of 50-100keV_{ee} were used to calculate standard PSD figure-of-merit, FoM, according to

$$FoM = \frac{\langle n \rangle - \langle \gamma \rangle}{\Gamma_{FWHMn} + \Gamma_{FWHM\gamma}}, \quad (3)$$

112 where $\langle n \rangle$, $\langle \gamma \rangle$, $\Gamma_{FWHMn}=2.35\sigma_n$ and $\Gamma_{FWHM\gamma}=2.35\sigma_\gamma$ represent centroids
 113 and full-widths-at-half-maximum of Gaussian fits to the PSD distributions.
 114 Errors were calculated from propagation of parameter errors returned by the
 115 fitter.

116 Theoretical estimates of the performance of various ^{10}B -loadings have
 117 been obtained by performing MCNPX simulations using Neutron Capture
 118 Ion Algorithm (NCIA). All simulations have been carried out for 10^5 events,
 119 exposing scintillators with various concentrations of ^{10}B and thicknesses to
 120 thermal neutron flux (25meV). Neutron source of the size of scintillator area
 121 was set up with neutron initial direction normal to the detector face. Param-
 122 eters effecting detector size, efficiency and resolving power, such as capture
 123 efficiency and average capture time, have been investigated across a range
 124 of ^{10}B concentrations. Histories were printed out on an event-by-event
 125 basis using ptrac card, and an algorithm was developed to post-process the
 126 ptrac file.

127 In order to calculate capture time, the time difference between thermal
 128 neutron injection into scintillator and alpha particle emission upon neutron
 129 capture were used to generate capture time distribution. The average capture
 130 time was calculated by performing an exponential fit to the distribution.

131 Thermal neutron capture efficiency was calculated using pulse height tally
 132 (f8) to group events in energy bins for alpha particles produced in the capture
 133 reaction, ensuring that the neutron is captured and alpha particle deposits
 134 all of its energy to the scintillator.

135 **3. Results and Discussion**

136 *3.1. Pulse Shape Discrimination*

137 Transparent, colorless samples (see Fig. 1) of PSD plastic scintillators
 138 containing from 0.4 to 5 wt.% of m-carborane with natural boron abundance
 139 were produced. The concentrations used (0.4; 0.75; 2.0; and 5.0 wt.%) corre-
 140 spond to ^{10}B content of (0.06; 0.11; 0.30; and 0.74 wt.%). Their performance
 141 in terms of light collection efficiency and pulse shape discrimination was stud-
 142 ied in comparison to a control sample, a PSD plastic prepared under the
 143 same conditions without boron.

144 In order to identify scintillator signatures specific to the type of radi-
 145 ation, energy calibrated pulse shape discrimination (PSD) is illustrated in
 146 Fig. 2 a) and b) for a PSD plastic scintillator and its loaded analogue
 147 containing 5wt.% of m-carborane, respectively. The PSD plastic exhibits



Figure 1: PSD plastic scintillator samples containing 5wt.% of m-carborane with natural boron abundance.

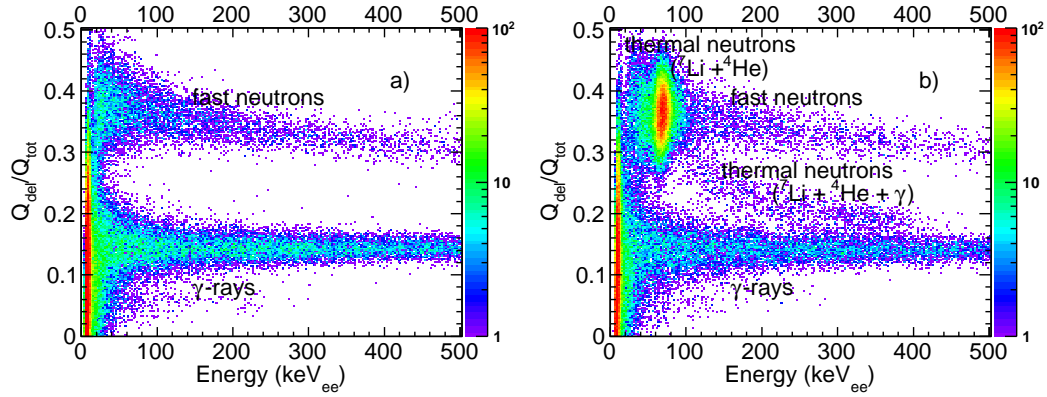


Figure 2: Pulse shape discrimination vs energy in electron equivalent units for a) PSD plastic exposed to neutron flux from 15- μ Ci ^{252}Cf source shielded with 7.5-cm of high density polyethylene (HDPE) and 5-cm of lead, b) plastic containing 5wt.% of m-carborane with natural abundance of boron under the same experimental conditions. Intensities, normalized with respect to time and solid angle coverage, can be used for efficiency comparison. Color scale represents counts.

148 typical response to γ -rays and fast neutrons, illustrated by two bands. The
 149 lower band, characteristic of smaller amount of delayed light, represents γ -
 150 ray events (scattered electrons). The upper band, with about 30-40% higher
 151 amount of the delayed light, exhibits a pattern characteristic to fast neu-
 152 trons (recoiled protons). PSD plastic containing boron provides a response
 153 characteristic to thermal neutrons in addition to the typical signatures of γ -
 154 rays and fast neutrons. It should be noted that in thermal neutron capture
 155 reaction on ^{10}B , two branches are expected according to Eq. 1 giving rise
 156 to two different signatures. The first type corresponds to energy deposition
 157 by $^7\text{Li}+^4\text{He}$. Because of constant energy of the charged reaction products
 158 defined by reaction kinematics and their short range of interaction, light out-
 159 put and its delayed fraction is well reproducible as illustrated by a signature
 160 with a mean of $72.6 \pm 0.01 \text{ keV}_{ee}$ and 0.387 ± 0.0003 in energy and PSD in Fig.
 161 2 b), respectively. The second type of thermal neutron signature represents
 162 events characteristic to energy deposition of $^7\text{Li}+^4\text{He}$ and a capture γ -ray.
 163 Incident γ -rays on organic scintillator interact mainly by Compton scatter-
 164 ing, and their energy deposition depends on scattering angle with maximum
 165 characteristic to Compton edge (313keV for 480keV capture γ -ray). Since
 166 scintillator response time is slower than γ -ray transit time, light output is a
 167 convolution due to excitation by charged reaction products and the capture
 168 γ -ray. Both, total light output and its delayed fraction varies depending on
 169 the capture γ -ray energy deposition. The ratio of delayed light is decreasing
 170 as the energy deposition is dominated by the capture γ -ray. Therefore, the
 171 second signature, characteristic to thermal neutrons, is shown as a band ex-
 172 tending towards typical γ -ray signature. Its maximum energy corresponds
 173 to the sum of capture γ -ray Compton Edge energy and total energy deposi-
 174 tion by charged reaction products (see Fig. 2 b)). Although 480keV γ -ray is
 175 emitted with 94% probability, the vast majority of the capture γ -rays escapes
 176 without depositing any energy to the scintillator due to the small sample size
 177 (2.5-cm diameter and 1-cm thickness), which is consistent with the two or-
 178 ders of magnitude higher intensity observed in the thermal neutron spot than
 179 in the band.

180 PSD distributions in the energy range 50-100keV $_{ee}$ are illustrated in Fig.
 181 3 a) and b) for a PSD plastic and a B-loaded PSD plastic, respectively,
 182 to show the degree of neutron- γ separation and effect of boron loading on
 183 neutron detection efficiency. For each sample in Fig. 3 distributions are
 184 shown with and without external moderator (Pb+HDPE, and only Pb, re-
 185 spectively). These results indicate comparable scintillator performance in

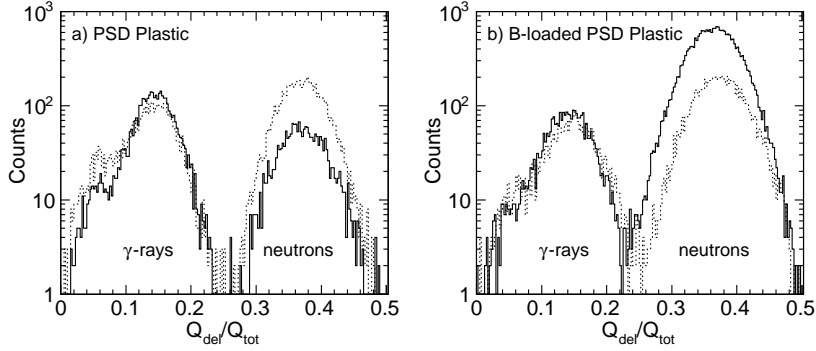


Figure 3: PSD distributions in energy range 50-100keV_{ee} corresponding to data illustrated in Fig. 2 for a) unloaded PSD plastic, and b) PSD plastic containing 5wt.% of m-carborane with natural boron abundance. Dotted line distributions, obtained for samples exposed to 15- μ Ci ^{252}Cf source without additional moderator, show comparable intensity for both samples. Distributions represented by full lines, obtained with 7.5-cm of moderator (HDPE), illustrate significant increase in neutron counts for boron-loaded plastic compared to PSD plastic sensitive only to fast neutrons. Integrals, normalized with respect to time and solid angle coverage, can be used for efficiency comparison.

186 the absence of thermal neutrons, and significant gain in neutron detection
 187 capability in the case of boron-loaded PSD plastic as compared to unloaded
 188 PSD plastic exposed to moderated neutron flux. Figure 4 illustrates neutron-
 189 γ discrimination performance of boron-loaded plastic scintillator in 100keV_{ee}
 190 energy bins for a range of energies from 100 to 500keV_{ee}. The distributions
 191 show clear separation between fast neutrons and γ -rays but partial overlap
 192 between thermal neutrons and γ -rays for events where the capture γ -ray
 193 undergoes scattering in the scintillator.

194 Additional studies were conducted on boron-loaded liquid scintillators
 195 to investigate PSD limits. The liquid scintillators were used for research
 196 purposes because of lower cost, easiness and short lead time for preparation of
 197 various compositions. Results are shown in Figs. 5-6 for xylene-based liquid
 198 scintillator containing 1wt.% of DPA (9,10-diphenylanthracene) and 1wt.%
 199 of boron oxide (III), enriched in ^{10}B , dissolved in 20wt.% of methanol). PSD
 200 figure-of-merit in energy range characteristic to thermal neutrons (see Fig. 6
 201 a)), 50-100keV_{ee}, is 1.67 ± 0.01 which is 20% higher than for typical plastics
 202 in the same energy range (1.4 ± 0.03). The results also indicate that the clear
 203 signal due to $^7\text{Li} + ^4\text{He}$ and a capture γ -ray could help better estimate the
 204 total number of neutron capture events statistically. It also should be noted

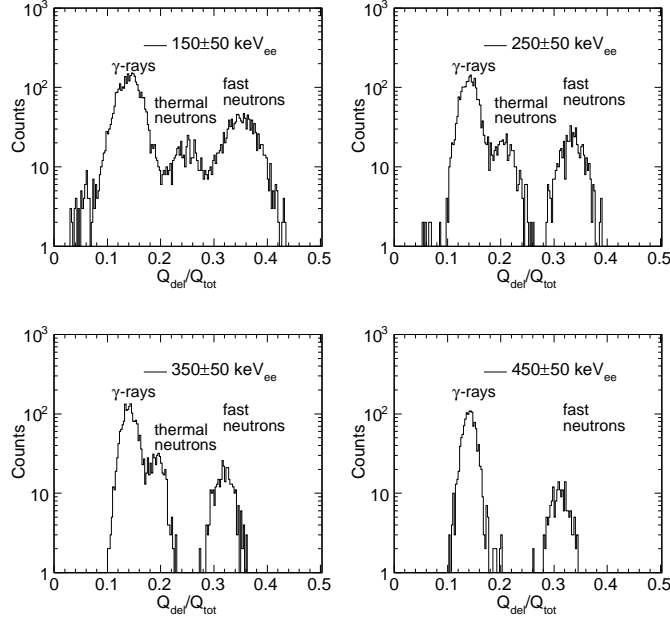


Figure 4: PSD distributions in several energy bins for PSD plastic containing 5wt.% of m-carborane. A sample of 2.5-cm diameter and 1-cm thickness was exposed to neutron flux from 15- μ Ci ^{252}Cf source shielded with 7.5-cm of high density polyethylene (HDPE) and 5-cm of lead.

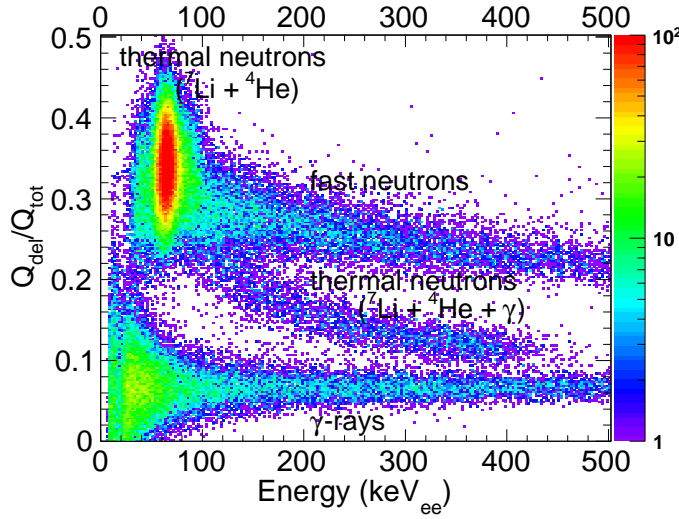


Figure 5: Pulse shape discrimination vs energy in electron equivalent units for a xylene-based liquid scintillator containing 1wt.% of DPA (9,10-diphenylanthracene) and 1wt.% of boron oxide (III) dissolved in 20wt.% of methanol. A sample of 5-cm diameter and 2.5-cm thickness was exposed to neutron flux from 15- μ Ci ^{252}Cf source shielded with 7.5-cm of high density polyethylene (HDPE) and 5-cm of lead. Color scale represents counts.

205 that the liquid scintillator provides a significantly better discrimination level
 206 than the currently available commercial scintillators [12, 13, 14, 15].

207 3.2. Effects of boron concentration

208 Incorporation of m-carborane into the scintillator requires careful under-
 209 standing of effects of a boron-containing compound on the scintillator light
 210 collection and pulse shape discrimination properties. Therefore, plastic scin-
 211 tillator development was performed by balancing neutron detection efficiency,
 212 time properties and light yield in connection with ^{10}B concentration.

213 Light yield dependence measured in response to ^{137}Cs source on m-carborane
 214 concentration relative to a control sample shows a moderate decrease as il-
 215 lustrated in Fig. 7 a). A decrease of 20% in the light yield for a PSD plastic
 216 loaded with 5wt.% of m-carborane was found. However, it is the light yield
 217 generated by capture reaction products that is of particular interest for the
 218 scintillator under development. It is needed to estimate if there are additional
 219 light losses due to quenching in presence of reaction products. Therefore, the
 220 capture reaction products light yield was obtained by performing Gaussian

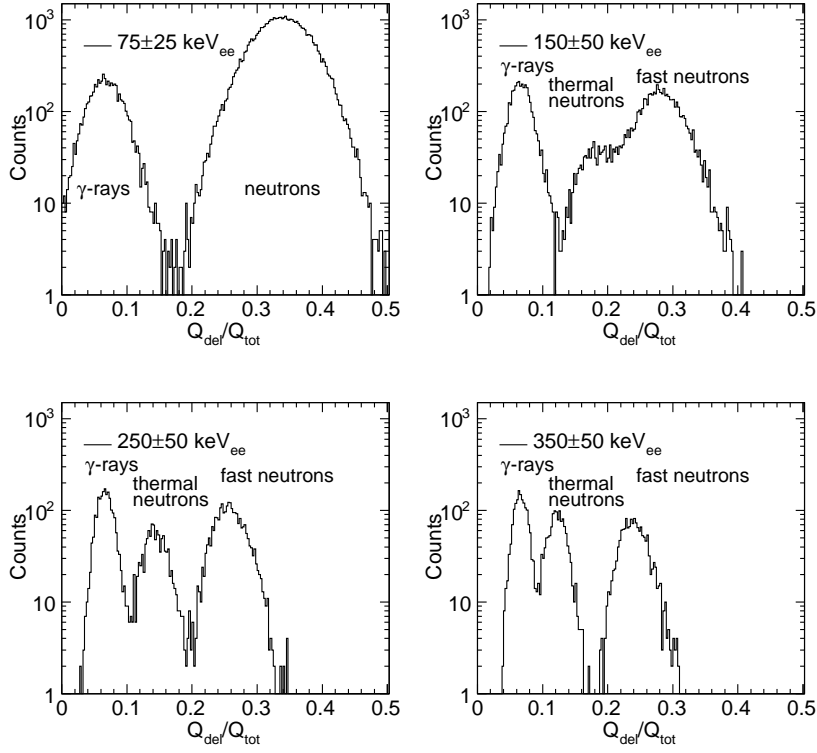


Figure 6: PSD distributions in several energy bins obtained with xylene-based liquid scintillator containing 1wt.% of DPA (9,10-diphenylanthracene) and 1wt.% of boron oxide (III), enriched in ^{10}B , dissolved in 20wt.% of methanol. A sample of 5-cm diameter and 2.5-cm thickness was exposed to neutron flux from 15- μCi ^{252}Cf source shielded with 7.5-cm of high density polyethylene (HDPE) and 5-cm of lead.

fits to the charge deposition characteristic to thermal neutron capture signature in pulse integral spectra. As a reference PSD plastic with 0.75wt.% of m-carborane loading was used. The results were compared to data obtained with ^{137}Cs source in Fig. 7 b). No additional losses were found for the capture reaction products, which is consistent with reduced light transmission due to attenuation rather than quenching.

Effects of m-carborane loading on neutron- γ discrimination properties, separation and figure-of-merit (FoM) values have been studied in energy domain characteristic to thermal neutrons 50-100keV $_{ee}$. Results illustrated in Fig. 7 c) show that separation remains constant across the range of m-carborane concentrations, which indicate that m-carborane does not affect the PSD process. Figure-of-merit values, on the other hand, slightly decrease with increasing m-carborane concentration, from 1.6 without boron loading to 1.4 with 5wt.% of m-carborane, respectively. The reason behind the decrease in the FoM is widening of neutron and γ PSD distributions due to light losses. For that purpose, the trend for full widths at half maximum, Γ_{FWHM} , is plotted in Fig. 7 d). This result is consistent with previous finding of moderately increasing light loss with increasing m-carborane concentration.

3.3. Simulation Predictions

Scintillator experimental development was combined with Monte Carlo calculations to estimate target ^{10}B concentration based on capture time and thermal neutron capture efficiency. Results of these calculations are presented in Figs. 8-10. It should be noted that the results of theoretical predictions obtained with MCNPX will be affected by scintillator neutron- γ discrimination levels which depend on many factors, including but not limited to count rate, scintillator size, setup components and their operation conditions; pmt, base, digitizer, sample rate, cable length [19], and should be evaluated experimentally as a part of calibration.

Capture time has been calculated as the time difference between thermal neutron injection into scintillator and the neutron capture. Calculated average capture times, $\langle t_c \rangle$ indicate a decrease of $\langle t_c \rangle$ from 10.2 ± 0.03 to $0.9 \pm 0.002 \mu\text{s}$ with increasing ^{10}B concentration from 0.1 to 2.0 wt.%. A slow response can be desirable at high incident neutron count rates, however; the detector will be more susceptible to accidental background. On the other hand, a faster detector response implies a shorter diffusion delay of neutron capture and is more desirable for applications with low neutron count rates such as the search for illicit trafficking of SNM. In addition, scintillator decay

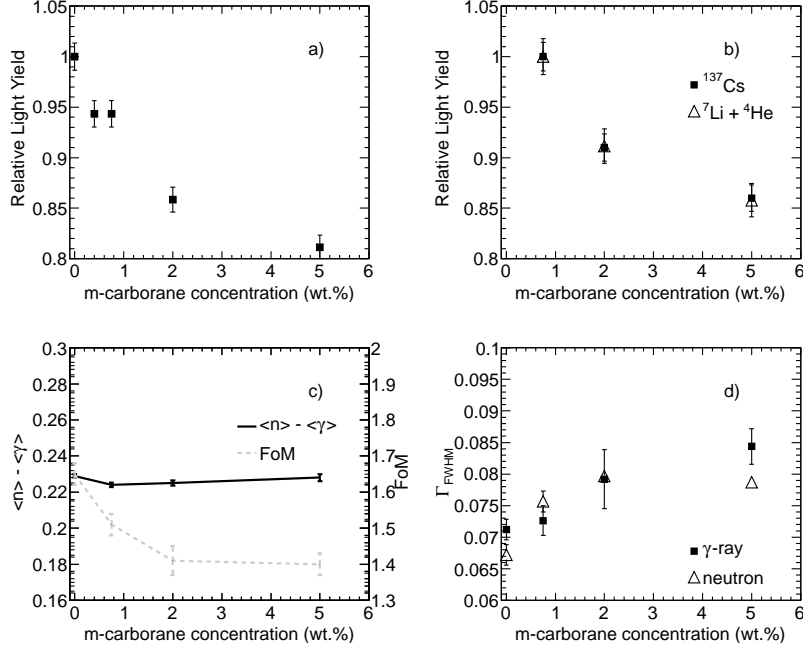


Figure 7: a) Light yield of PSD plastic samples containing various concentrations of m-carborane with natural boron abundance relative to a blank sample (PSD plastic), measured for ^{137}Cs . b) Light yield of PSD plastic samples loaded with various concentrations of natural m-carborane relative to a PSD plastic loaded with 0.75wt.% determined for thermal neutron capture reaction products. Comparison to the results obtained for ^{137}Cs source shows no additional light loss for capture reaction products. c) Neutron- γ separation and PSD figure-of-merit (FoM) vs m-carborane concentration. Separation is not influenced by m-carborane concentration while FoM shows slight decrease with increasing concentration due to moderate light loss. Lines are included to guide the eye. d) Full Width at Half Maximum (Γ_{FWHM}) of neutron and γ PSD distributions in energy range 50-100keV_{ee} vs m-carborane concentration.

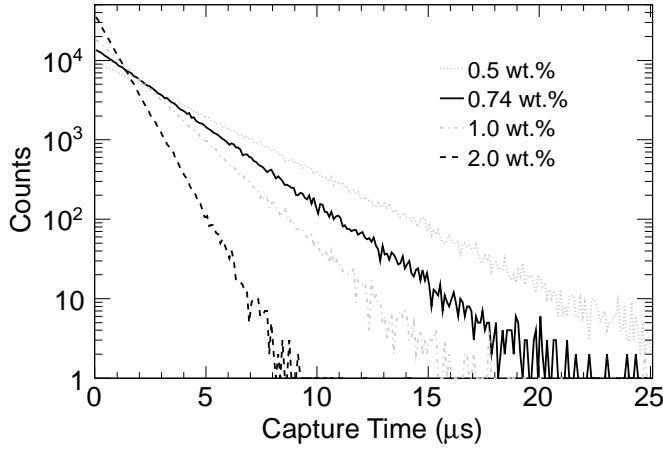


Figure 8: Simulated capture time distributions for PSD plastic containing concentrations of 0.5, 0.74, 1.0, and 2.0 of ^{10}B by weight.

characteristics should also be taken into consideration, particularly because in response to neutrons (thermal and fast) the PSD plastic scintillator emits a significant amount of delayed light required for effective PSD for approximately $1.3\mu\text{s}$.

Simulated thermal neutron capture efficiency on ^{10}B , presented in Fig. 10, increases with scintillator thickness to reach saturation. Both, the slope and the efficiency at saturation depend on ^{10}B concentration. With increasing ^{10}B concentration, capture efficiency approaches, but does not reach, unity mainly due to thermal neutron escape at detector boundaries. For the considered scintillator sizes, only marginal losses were found due to competing capture reaction on ^1H nucleus. B-loaded PSD plastic scintillator response to the 2.2 MeV γ -rays, emitted upon thermal neutron capture on ^1H nucleus, will essentially result in a pulse with decay characteristic to γ -rays (recoiled electrons). Therefore, these thermal neutron capture events cannot be discriminated directly against background γ -rays. Generally, larger ^{10}B concentrations will provide smaller size, more efficient detectors with faster response to thermal neutrons. However, all the parameters depending on ^{10}B mentioned above need to be optimized for specific conditions and applications.

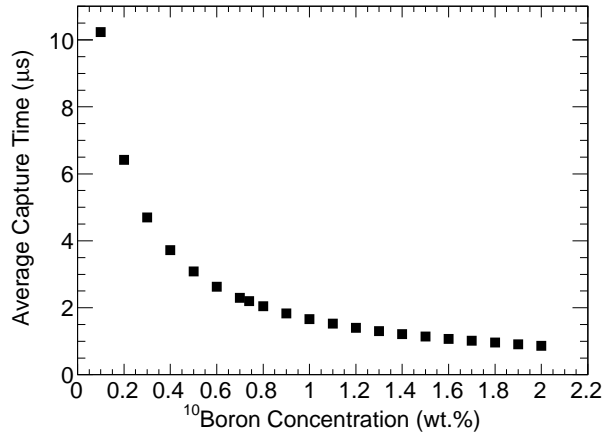


Figure 9: Simulated average neutron capture time vs ^{10}B concentration by weight. Error bars are within the points.

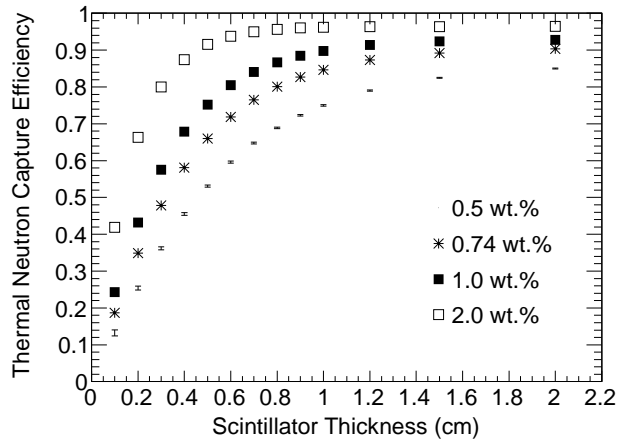


Figure 10: Simulated thermal neutron capture efficiency of ^{10}B vs scintillator thickness for ^{10}B loading of 0.5, 0.74, 1.0, and 2.0 % by weight. Error bars are within the points.

277 4. Conclusions

278 Development of pulse shape discriminating plastic scintillator for efficient
279 neutron detection has been demonstrated by incorporation of m-carborane.
280 The material introduces detector sensitivity to neutrons which are below
281 the threshold for detection by proton recoil maintaining sensitivity to fast
282 neutrons, while discriminating against γ -rays. This may be particularly im-
283 portant for passive detection of low flux neutron sources such as illicit nu-
284 clear material, where fission neutrons lose energy by scattering in shielding
285 and the environment before entering the detector. Simulation results sug-
286 gest that at 0.74% of ^{10}B loading thermal neutrons can be captured, on
287 average, within $2.2\mu\text{s}$ with detection efficiency of 84% in 1-cm thick scintilla-
288 tor. Incorporation of 0.74wt.% of ^{10}B in the form of carborane with natural
289 boron abundance requires 5% of carborane by weight, and does not influence
290 the scintillator pulse shape discrimination properties beyond a moderate de-
291 crease of light yield. A PSD figure-of-merit value of 1.4 ± 0.03 in the energy
292 range of 50-100keV_{ee} was obtained for PSD plastic loaded with 5wt.% of
293 m-carborane by integration of the delayed light from 45ns to 1.25 μs from
294 maximum. A combination of experimental and simulation results suggest
295 that boron-loaded PSD plastic is a good candidate for an efficient compact
296 neutron detection system.

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